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The thermal decomposition of TAGN is of interest for understanding its stability and compatibility with other nitramine propellant ingredients. Low vulnerability (LOVA) propellants containing inert polymers such as hydroxy-terminated polybutadiene or the cellulose acetate polymers and the nitramines HMX or RDX have been formulated at ICT. We are interested in determining the effect of TAGN on the stability of these propellants by studying their thermal decomposition at temperatures below the deflagration temperature (<220°C).

In this paper the preliminary results for this study are presented. Techniques used include thermal decomposition/MS, in which the evolved gases are analyzed by mass spectroscopy. The solid residue was analyzed by Fourier transform infrared spectroscopy (FTIR), elemental (carbon, hydrogen and nitrogen) analysis, thin layer chromatography, differential scanning calorimetry and atomic absorption spectroscopy. The results from the analyses agree and

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indicate that ammonia and nitrogen are the chief gases evolved and that the residue remains enriched in nitrate.

We have also investigated the difference between pink and pure TAGN crystals using FTIR spectroscopy. Although infrared spectroscopy had been previously used to study TAGN, no differences in the pink and pure samples were seen. In this work, pink TAGN and red crystals recovered from recrystallization of the pink material suggest that the pink TAGN is enriched with the nitrated functionality. The identity of this nitrate product is under investigation.

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# DECOMPOSITION AND STABILITY STUDIES OF TAGN

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by PAMELA J. KASTE and others
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DECEMBER 1988

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U.S. ARMY LABORATORY COMMAND

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## I. INTRODUCTION

Triaminoguanidium nitrate (TAGN) is of interest for a variety of RDX-based propellant applications such as low vulnerability (LOVA) propellant materials, gas generators and very high burning rate formulation. Its advantages include low molecular weight gas products and therefore low flame temperatures, as well as an increased muzzle velocity and impetus. Several reports have been published in which these advantages have been realized in ballistic measurements.[1-4] However, one critical disadvantage is the instability of TAGN, both alone and in propellant formulations. Colored (particularly pink) crystals of TAGN are commonly formed in the synthesis, storage and/or processing of TAGN. Care must be taken to avoid metal ion contamination and to ensure prompt solvent removal (particularly ethyl acetate, acetone, alcohols and those which are basic). However, even when such precautions are taken, the resultant TAGN often becomes discolored and problems with the propellants made from it are likely. In this laboratory differences in the physical appearance of propellants synthesized from pure TAGN versus an impure, pink TAGN were seen; a decrease in ballistic properties was also observed.[5] It has also been found that the addition of NC to propellant formulations can reduce the stability of TAGN propellants and that the presence of a stabilizer such as ascardite can minimize the loss of stability in NC/TAGN formulations.[6]

The purpose of this research was to study the decomposition of TAGN alone, as well as its effect on the family of cellulose acetate polymers. LOVA propellants consisting of cellulose acetate, RDX and TAGN have been produced and studied at ICT; therefore, the stability and compatibility of these propellant materials is of interest.

## II. EXPERIMENTAL

TAGN was studied at temperatures below its deflagration point (227°C), both alone or in cellulose acetate mixtures using mass spectroscopy (MS). The sample was thermally decomposed in a controlled temperature bath and was connected to the MS vacuum chamber via a heated transfer line and valve. The evolved gases were quantitatively measured as a function of temperature while the non-volatile residue was analyzed by methods described below. The sample was placed in the bath at 150°C and heated at approximately 10°C/min to its final temperature, nominally between 190-220°C. The sample was held at the final temperature for 15 min and the gases collected in the vacuum chamber; after that time the high resolution mass spectrum was obtained. The temperature of the transfer lines and vacuum chamber were kept high enough so that species such as $NH_3$ and acetic acid did not condense.

Infrared spectra were obtained using a 20SX Nicolet Fourier transform infrared (FTIR) spectrometer with MCT detector. One hundred scans at nominally two wavenumber resolution were signal averaged. A Perkin Elmer 300 series atomic absorption spectrometer was used for the determination of metals. The standard conditions suggested in the instrument manual were used for the selection of source lamp, flame gas and detection wavelength. Under these conditions, detection limits (in ppm) were as follows: Fe (0.005), Cu (0.1), Ni (0.004), Zn (0.001), Cd (0.001), Cr (0.001).

Differential scanning calorimetry (DSC) was performed at a heating rate of 10°C/min using a Mettler 3000 Thermal Analysis system. Nominal 3 mg samples were place into 4 mm diameter pans with crimped lids and a vent hole. The flow rate of gas over the sample was nominally 20 cc/min. TAGN and its decomposition products were determined qualitatively by thin layer chromatography (TLC). The best conditions found for their separation were silica TLC plates with acetone: 20% $NH_4OH$ (98:2) as the eluting solvent.

## III. THERMAL DECOMPOSITION/MASS SPECTROMETRIC ANALYSIS

The results for TAGN over the temperature range 190-220°C were rather similar, with ammonia (35-45%) and nitrogen (50-65%) as the chief products (Table 1). Other gases found in small amounts include CO (<2%), formaldehyde (<4%), NO (<1%), and possibly hydrazine (<2%). $N_2O$ was found in small amounts (<1%), usually only at the higher temperatures (210-220°C), or if prolonged (30-60 min) heating periods were used. Unfortunately, no difference was seen between the gases evolved from the pink and pure TAGN.

Table 1. Mass Spectrometric Analysis of the Gaseous Decomposition Products of TAGN Alone and Mixtures with Cellulose Acetate Binders

| | $NH_3$ | $N_2$ | CO | $CO_2$ | $H_2C{=}O$ | NO | $N_2O$ |
|---|---|---|---|---|---|---|---|
| TAGN | 35. | 58. | <2 | ---- | <4 | <1 | ---- |
| CA | | | | 100. | | | |
| TAGN/CA 75/25 | 3.8 | 58.8 | 3.6 | 16.7 | --- | 5.6 | 10.6 |
| TAGN/CAB | 17.9 | 62.4 | 1.3 | 5.8 | --- | 2.0 | 3.0 |

Note: $H_2O$ was not included in the above analyses.
Decomposition temperature was 210-220°C.
Values are in mole percent.

Since the interaction between TAGN and cellulosic binders is of interest, a mixture of TAGN:CA was also studied in the MS under the conditions mentioned above. Cellulose acetate (CA) alone yielded no permanent gases at temperatures up to 220°C; only acetic acid was formed. However, the 1:1 by weight mixture yielded almost no $NH_3$ or acetic acid, but gave copious amounts of $N_2O$. Mixtures of TAGN with cellulose acetate butyrate (CAB) demonstrated the same effects; $N_2O$ was observed in these mixtures as well. The $NH_3$ concentration in the mixture was lower than in the TAGN sample alone as well. The effect of $N_2O$ production and decreased $NH_3$ level is less with CAB than with CA, presumably due to the lower acetate substitution in the CAB.

Although it was suspected that the $NH_3$ produced from TAGN might react with CA, it was surprising to find that so much ammonia was consumed (assuming that is was formed, as in TAGN alone) and that such a large quantity of $N_2O$ was produced. In an attempt to determine what reactions might be involved in

the $N_2O$ production, a mixture of TAGN and underivatized cellulose (no acetate groups present) was decomposed in the same manner. The gases evolved consisted chiefly of ammonia and nitrogen, similar to the case of TAGN alone. Thus, it appears that the acetate groups are critical in the interaction between TAGN and CA in which $N_2O$ is produced in the CAB.

Further experiments were performed to determine if ammonia alone is sufficient to react with cellulose acetate and produce $N_2O$. An ammonium salt ($NH_4HCO_3$, which volatilizes at the 220°C used in these thermal decomposition to yield $NH_3$ and $CO_2$) was mixed with cellulose acetate and treated as above. No $N_2O$ was obtained. Conversely, a mixture of TAGN and acetic acid was prepared to determine if acetic acid is sufficient to react with TAGN and cause the formation of $N_2O$. Again, none was seen, the chief products being those of the individual components alone.

From these results we can conclude that the acetate functional group of cellulose acetate is essential for the reaction with TAGN to produce $N_2O$. It is not yet certain in what manner the TAGN molecule reacts with CA in this decomposition. Further experiments are planned to determine this as well as whether the reactions occur in the gas or condensed phases.

## IV. FTIR RESULTS

The infrared experiments were performed (a) in order to characterize the solid residue from the TAGN decomposition-MS experiments and (b) to determine any difference in a pure sample of TAGN and a batch of pink TAGN which was prepared at ICT. By analyzing both the solid and gaseous products from the MS thermal treatment, the decomposition mechanism of TAGN under the conditions used could be better elucidated.

TAGN samples were heated for MS decomposition studies as described above. Since $NH_3$ is the chief decomposition gas it was quite logical to suspect that the less substituted guanidine nitrates might be formed in the decomposition of TAGN. Thus, the spectra of diaminoguanidine nitrate (DAGN), aminoguanidine nitrate (AGN), and guanidine nitrate (GN) as well as TAGN were obtained as reference compounds. These are shown in Figure 1 and the absorption frequencies and assignments of the major bands are shown in Table 2. The infrared spectra and band assignments for TAGN had previously been reported by Brill.[7] Some features of the reference spectra are worth noting. As the NH groups are removed from TAGN, the bands due to NH asymmetric stretch begin to broaden. The C-N band at 1685 in TAGN (with partial double bond character) shifts to lower frequency with fewer NH groups.

The N-N band of medium intensity at 1129 $cm^{-1}$ in the TAGN spectrum has a significantly lower relative intensity in DAGN and is barely discernable in AGN. This is expected, based on the structure of these compounds. The N-N band is fairly weak, even in TAGN which has three N-N bonds. DAGN has one less N-N bond and the band is weaker than in TAGN. In AGN (only one N-N bond) the band is barely discernable. The other prominent bands in TAGN are the nitrate bands, the strongest of which is at 1384 $cm^{-1}$ and a small but sharp band at 825 $cm^{-1}$. These bands were identified from a pure sample of $KNO_3$ and appear to be stronger in the decomposition products, relative to TAGN.

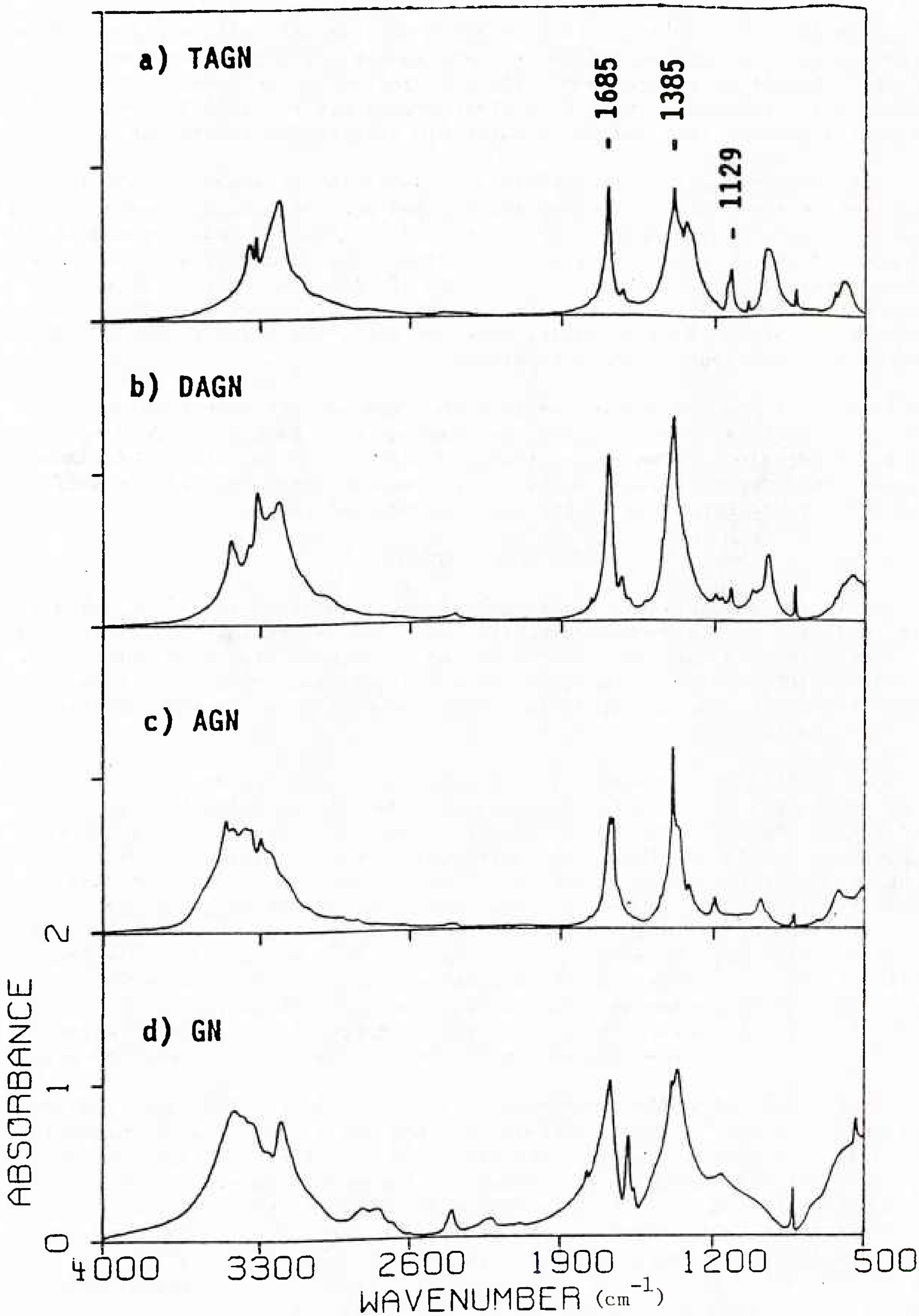


Figure 1. Infrared Spectra of the Tri-, Di-, Mono-Amino and Unsubstituted Guanidine Nitrates

Three samples were chosen from the MS experiments for analysis by IR, to determine what differences might be seen relative to the pure TAGN. Two samples were solid residues, obtained by heating TAGN at 210°C and 220°C for 15 minutes. The third sample consisted of a powder which was initially only slightly off-white in color and which was found condensed on the side of a glass tube in a small area where the glass was not completely wrapped with heating tape. This condensed material was removed from the tube (inadvertantly with a metal spatula) and saved in a glass vial. Two days later it was noticed that the material was brilliant red and was therefore of significant interest. The infrared spectra of this material is shown in Figure 2, along with that of DAGN; the similarity is striking and shows that the condensate consists chiefly of DAGN. The pink color is due either to an impurity or decomposition product; pure DAGN is a white crystalline material.

The solid decomposition residues are not so easily identified although trends in their spectra suggest general changes in the structure of the TAGN. The TAGN heated to 210 was originally light pink and yielded a gummy residue which was medium pink and whose spectrum is seen in Figure 3. A pure sample of TAGN was heated to 220°C and yielded a residue of similar texture to that at 220°C, but was only light pink in color. The absence of the N-N band suggest that most of these bonds have been broken; the broad and featureless NH bands also indicate increased decomposition. The residue probably consists of a mixture of products and thus, identification is difficult. However, it appears likely that TAGN forms such compounds in its early decomposition.

Table 2. Major Infrared Absorption Band Assignments for the Tri-, Di-, Mono-Amino and Unsubstituted Guanidine Nitrates

| IR | $NH_2$ | N-C-N | N-N |
|---|---|---|---|
| Sample: | | | |
| TAGN | 3214-3319 | 1685 | 1129 |
| DAGN | 3216-3353 | 1682 | 1127 |
| AGN | 3263-3452 | 1671/1657 | 1120 |
| GN | 3202-3448 | 1666 | ---- |
| Heated: | | | |
| Red Condensate | 3213-3319 | 1684 | 1128 |
| TAGN Residue (210°C) | 3215-3307 | 1660 | ---- |
| TAGN Residue (220°C) | 3157-3385 | 1654 | 1130 |
| Unheated: | | | |
| Red Crystals from Recrystall. Solvent | 3212-3319 | 1685 | 1128 |
| Pink TAGN | 3213-3343 | 1685 | 1128 |

Note: The nitrate bands have the same frequency for each sample and are at 1385 $cm^{-1}$ and 825 $cm^{-1}$.

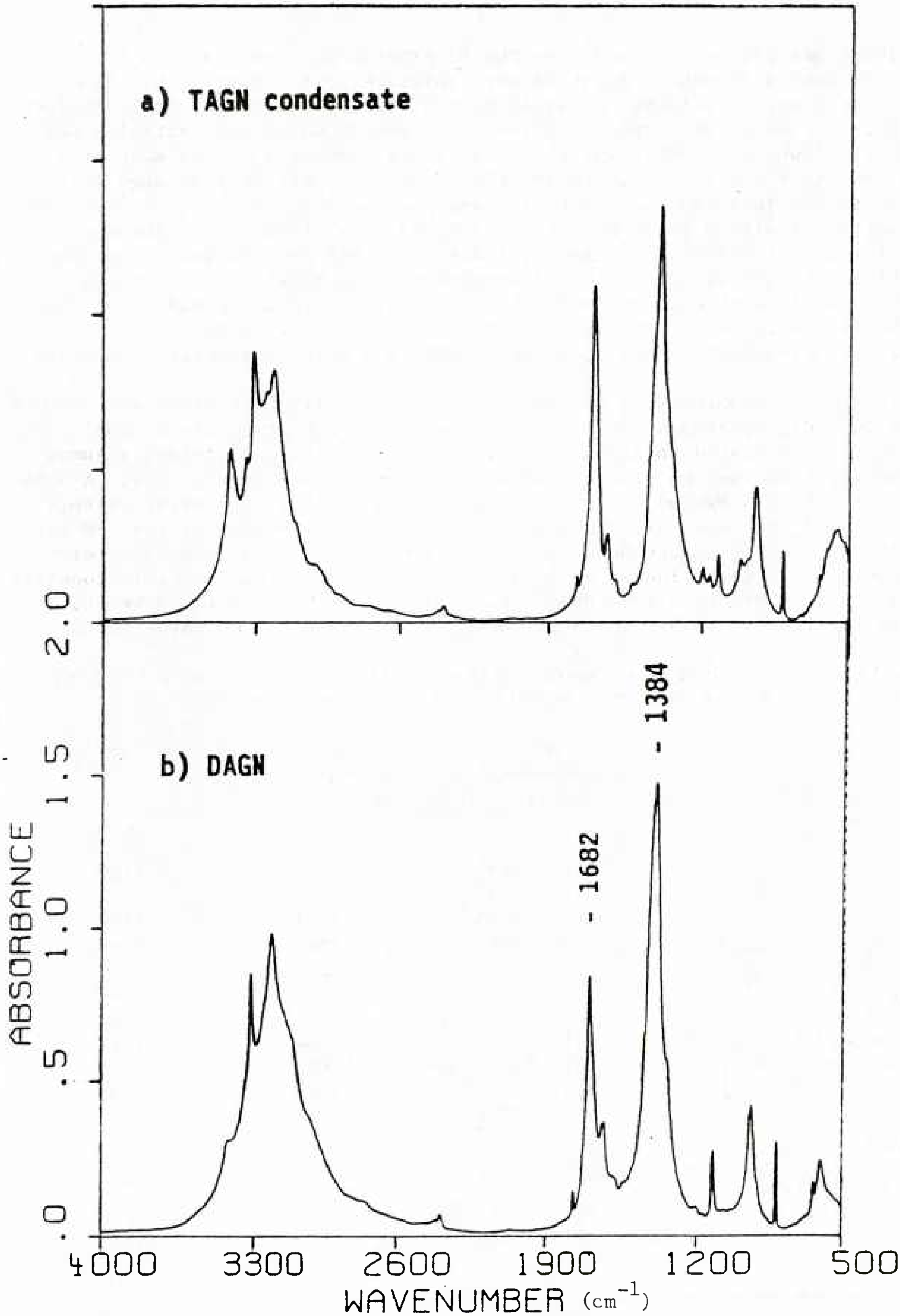


Figure 2. Comparison of the Infrared Spectra of a TAGN Decomposition/ Condensate Product and DAGN

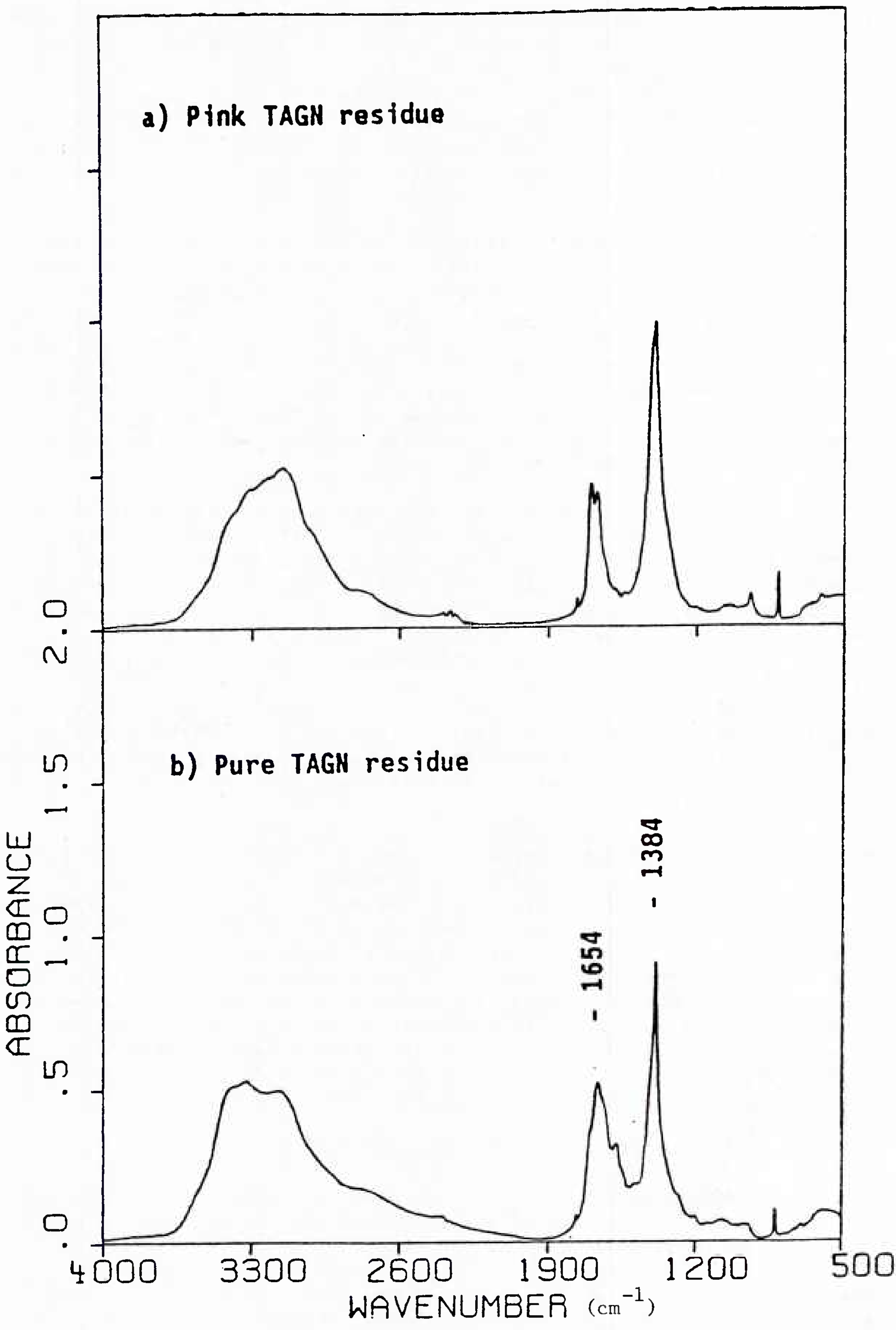


Figure 3. Infrared Spectra of the Solid Decomposition Products of TAGN Heated to 210-220°C

The next set of IR samples were analyzed to determine if any difference in the spectra of pink or pure TAGN samples could be determined. Previous researchers had detected no differences but it was decided to try again. A medium pink sample of TAGN (the pinkest available) was chosen, but was still damp from the mother liquor. Unfortunately, when this sample was dried for IR analysis, most of the pink color was also removed. A similar effect had also been previously reported.[8] The spectrum was similar to that of TAGN and is not shown.

It was known that the pink crystals can be washed with deionized water and the pink color removed. TAGN crystals appear as hollow cylinders when viewed under a microscope. It is believed that much of the pink color is due to liquid trapped in the hollow spaces of the cylinder.[9] At ICT researchers had also prepared a sample of crystals which, when washed, turned the deionized water a pink-orange color. This wash-water was collected and concentrated (under vacuum-no heat) for IR analysis. In the first attempt the sample was extracted into methylene chloride (which then turned pink) and evaporated to dryness. However, the resulting crystals were white and the spectrum was identical to TAGN (not shown).

Another attempt was made using this recrystallization liquor in which the water was again removed under vacuum without heat. After several hours under vacuum the solution remained clear, but turned a beautiful red color. The water was completely evaporated and the resulting crystals were run as KBr pellets and the results are shown in Figure 4a. The only difference between this and the TAGN spectra (Figure 4b) is the stronger nitrate band of the former. This might suggest that the sample becomes enriched in nitrate. If so, unlike the decomposition spectra discussed above, the larger nitrate band is not accompanied by any change in the TAG (triaminoguanidine) portion of the molecule; the band positions are similar to those of TAGN (Table 1). Thus, the pink color could be due to some nitrate complex (possibly a metal nitrate) present in TAGN which, except for the nitrate band, does not absorb significatly in the IR.

Previous researchers have speculated that the pink color is due to the formation of conjugated double-bonded complexes which form from TAGN after the loss of nitrate, and that the latter is washed away in the recrystallization fluid.[8] However, the nitrate band in the spectra of the pink and red crystals described above does not support this. Also, if such conjugated TAG-like complexes did form, they would expect to be at least weakly IR absorbing, being C=N and N=N type bands. Thus, in such deeply red crystals as used in this experiment one might expect to see at least minor changes in the IR spectrum. None were observed, except for the nitrate band intensity, as indicated above. Whether the stronger nitrate band is due to an increased concentration of nitrate or simply to crystal effects is not yet known.

## V. ATOMIC ABSORPTION ANALYSIS

Since it seemed possible that the pink (red) color of some the samples might be related to the presence of metal, atomic absorption spectroscopy (AAS) analyses were run on several of the samples.

The metals determined were those which are most common: Fe, Cu, Ni, Zn, Cd and Cr. The level of all of these was below their detection limits. The

likelihood of contact of the sample with any other metal is slim and thus, it seems doubtful that any residual metal complex impurity was the cause of the pink color.

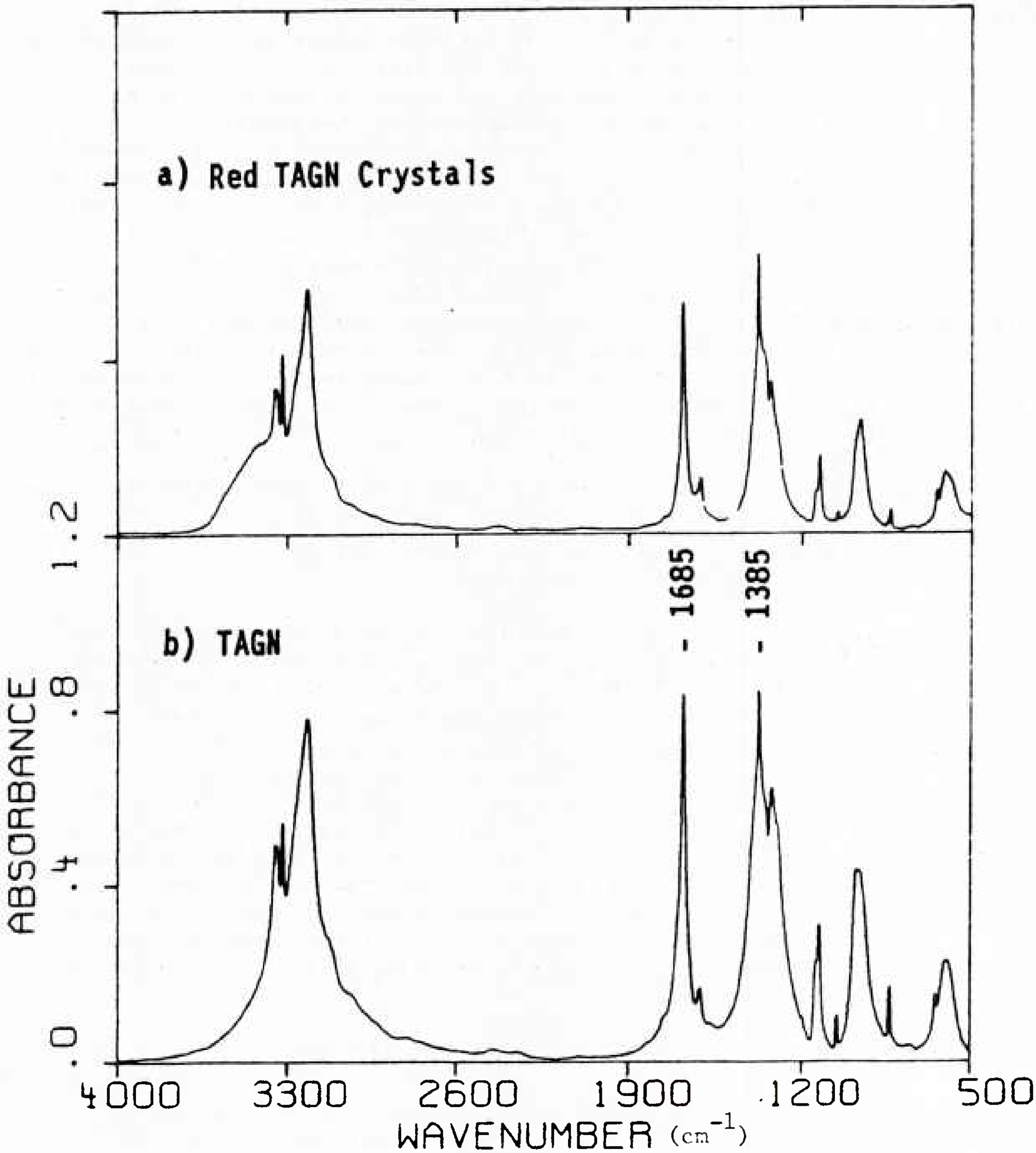


Figure 4. The Infrared Spectra of: (a) Deep Red Crystals Obtained by Vacuum Drying the Recrystallization Water and (b) TAGN

## VI. C, H, N, AND O ANALYSIS

Elemental analysis was performed on duplicate samples of pure TAGN, pink TAGN and crystals which formed from the recrystallization solvent (all samples were dryed under vacuum to ensure that any residual solvent was removed). Samples of the residue obtained by heating TAGN to 210 or 220°C for 15 min were also analyzed. Duplicate samples of the pure TAGN residue were prepared. Single samples of pink TAGN and pink TAGN which had been washed until it became white were also analyzed, as well as a single sample of the red-colored material which had condensed on the walls of the test tube while heating a pure sample. The results of these analyses are shown in Tables 3 and 4. In the case of the duplicated samples the average of the two results and the standard deviation is given (Table 2). The delta ($\Delta$) value is the difference between the theoretcial value for TAGN and the analytical value; the % value is the % change from the theoretical values for a sample of pure TAGN (Table 4). For the pure TAGN sample $\Delta$ gives an estimate of the accuracy of the analysis. It can be seen that the C and H values are more accurate than are those of nitrogen; those of oxygen are obtained by subtracting the sum of the others from 100 and are the least accurate. Results for nitrogen with the instrument used in this work are consistently less accurate then for carbon or hydrogen.[10] Whether this accounts for the discrepancy between the theoretical and unheated, pure TAGN in Table 4 is unclear. Thus, there may be some minor impurity/decomposition within the parent material.

Unfortunately, no significant difference was seen between either the pink TAGN crystals or the crystals obtained from the recrystallization fluid. However, the latter were only very slightly pink and are not the brilliant red crystals on which the IR analysis was made.

The results for the decomposition residue and, to a lesser extent, the condensate show that these samples are enriched in C and $O_2$ and have less $H_2$ and $N_2$ present, relative to TAGN. This is in agreement with the MS results which have indicated that the chief products are $N_2$ and $NH_3$, and that there are copious amounts of both formed. Other gases (containing C and $O_2$) are found in only small quantities at high temperature and/or longer heating durations. Since the only source of oxygen in the TAGN molecule is the $HNO_3$ group, the enrichment of $O_2$ supports the IR result (formation of DAGN and AGN as the sample decomposes) that decomposed samples are enriched in nitrate. Any enrichment of nitrogen due to the nitrate is negated by the large loss of $N_2$ and $NH_3$ and thus the percent nitrogen in the decomposed samples is lower than that of TAGN. It should be noted here again that the condensate and pink TAGN were only pale pink in color and were not the samples for which the IR results were obtained.

## VII. DIFFERENTIAL SCANNING CALORIMETRY

In the DSC at 10°C/min, the exothermic decomposition of TAGN begins immediately after the melting point and reaches its maximum at about 232°C (Figure 5). A second, slightly sharper exotherm is observed at 278°C, with a smaller and more variable exotherm (265°C) occurring between the 2 larger exotherms. The first exotherm (just after the melting point) is the one associated with the reactions studied in the mass spectrometer (mainly $N_2$ and $NH_3$ evolution). This is evidenced by the fact that when the residue from the MS experiments is run in the DSC, the first exotherm normally seen in TAGN is

not present in the curve for the residue (Figure 6), and only the second major exotherm is seen. In addition, this second peak occurs at the same temperature as that found when DAGN is decomposed in the DSC (Figure 6b). This lends further support to the MS, FTIR and elemental analyses that DAGN is formed from mild decomposition of TAGN.

Table 3. Elemental Analysis of TAGN and the Residue/Condensate from the MS Experiments; the Theoretical Value for Pure/Undecomposed TAGN is Given as a Reference

| Sample | C | H | N | O 100-(C+N+H) |
|---|---|---|---|---|
| TAGN (Theoretical) | 7.18 | 5.42 | 58.64 | 28.73 |
| Unheated: | | | | |
| TAGN (Pure) | 7.11 ± .05<br>Δ = − 0.07 | 5.45 ± .01<br>Δ = 0.03 | 55.75 ± .89<br>Δ = − 2.89 | 31.69 ± .83<br>Δ = − 2.96 |
| TAGN (Pink) | 7.20 ± .02<br>Δ = 0.02 | 5.46 ± .03<br>Δ = 0.04 | 55.44 ± .82<br>Δ = − 3.20 | 31.88 ± .85<br>Δ = 3.15 |
| Crystals from Recrystallization Solvent | 7.14 ± .01<br>Δ = − 0.04 | 5.51 ± .01<br>Δ = 0.09 | 55.77 ± .71<br>Δ = − 2.87 | 31.57 ± .71<br>Δ = 2.84 |
| Heated: | | | | |
| Pure TAGN Residue | 9.63 ± .20<br>Δ = 2.45 | 4.89 ± .02<br>Δ = − 0.50 | 42.88 ± .57<br>Δ = − 16.33 | 42.59 ± .38<br>Δ = 13.86 |
| Washed Pink TAGN Residue | 9.78<br>Δ = 36.20 | 4.51<br>Δ = − 0.91 | 41.49<br>Δ = − 17.15 | 44.22<br>Δ = 15.49 |
| Pink TAGN Residue | 11.99<br>Δ = 4.80 | 4.40<br>Δ = − 1.02 | 45.00<br>Δ = − 13.64 | 38.61<br>Δ = 9.88 |
| Condensate from Pure TAGN | 8.83<br>Δ = 1.65 | 4.94<br>Δ = − 0.48 | 49.84<br>Δ = − 8.80 | 36.39<br>Δ = 7.66 |

Note: Values with error bars are the average of two measurements. Delta (Δ) represents the difference between the experimental and theoretical values.

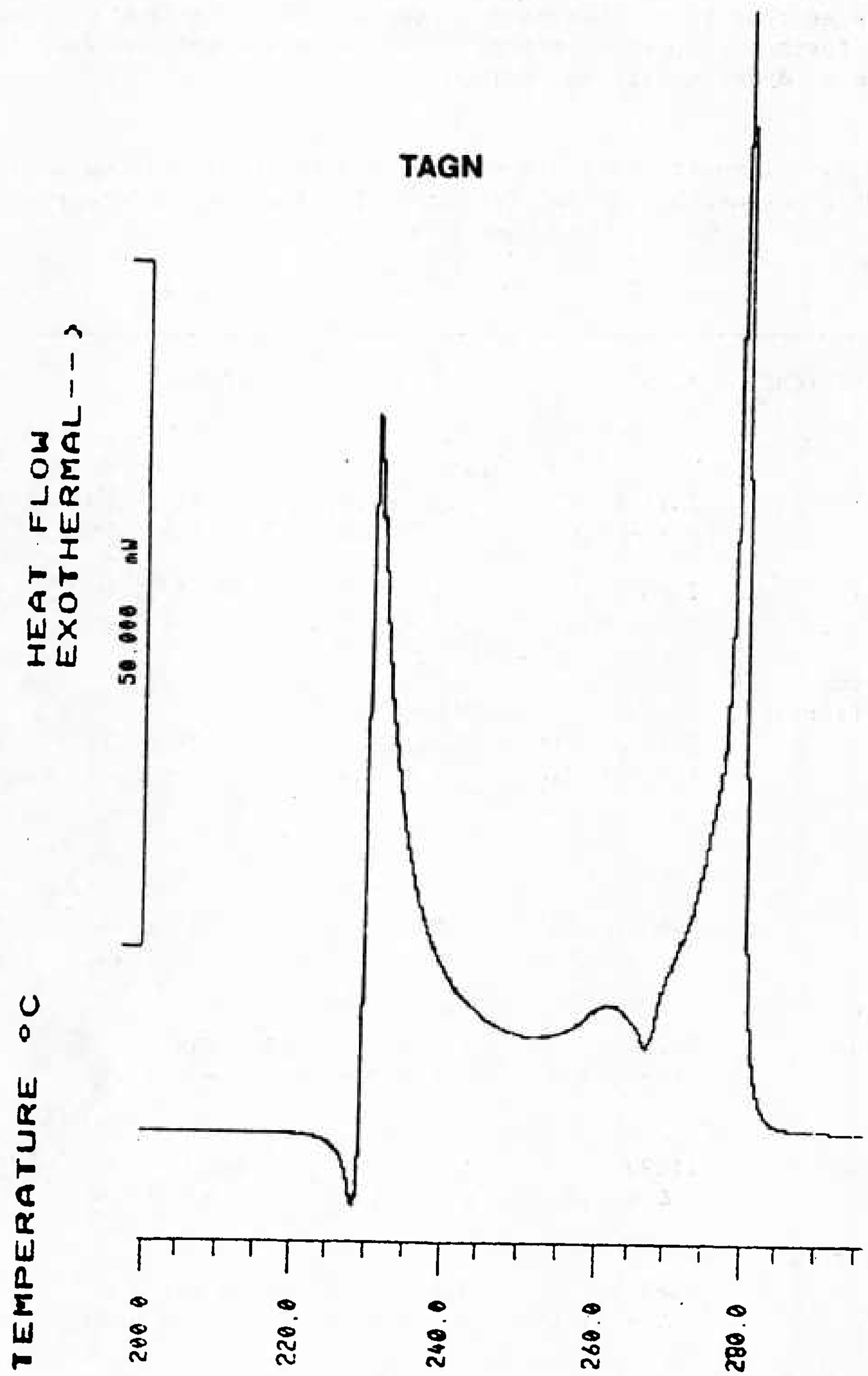


Figure 5. Differential Scanning Calorimetry Analysis of TAGN

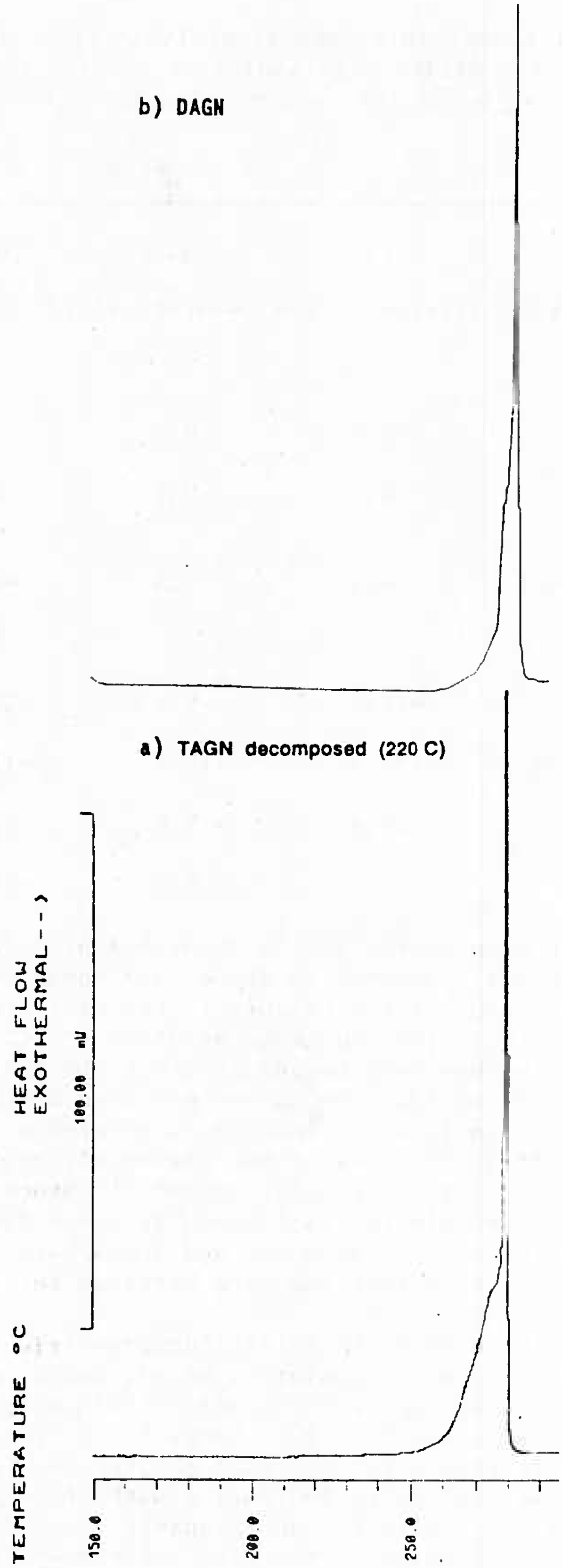


Figure 6. Differential Scanning Calorimetry Analysis of (a) the Solid Decomposition Residue of TAGN and (b) DAGN

Table 4. Percent Change in Elemental Analysis from the Theoretical Values for TAGN. A positive sign indicates a value greater than the theoretical; a negative value indicate a value less than the theoretical.

| Sample | C | H | N | O |
|---|---|---|---|---|
| TAGN (Theoretical) | 7.18 | 5.42 | 58.64 | 28.73 |
| Percent Difference from Theoretical Value: | | | | |
| Unheated: | | | | |
| TAGN (Pure) | -0.9 | 0.5 | 4.9 | -10.3 |
| TAGN (Pink) | 0.2 | 0.7 | 5.4 | 10.9 |
| Crystals from Recrystallization Solvent | -0.5 | 1.6 | -4.9 | 9.8 |
| Heated: | | | | |
| Pure TAGN Residue | +34.1 | -9.8 | -27.8 | +48. |
| Washed Pink TAGN Residue | +36.2 | -16.8 | -17.2 | +54.9 |
| Pink TAGN Residue | +66.8 | -18.8 | -23.2 | +34.1 |
| Condensate | +22.9 | -8.8 | -15.0 | +26.6 |

TAGN and RDX alone give what might be described as a "controlled reaction" (peaks are roughly Gaussian in shape) and occur over a range of several minutes. In the case of RDX (Figure 7) the exothermic reaction begins just after melting (at about 210) and peaks at about 235°C. However, when TAGN is mixed with RDX the exotherm begins at about 210 and reaches its maximum almost immediately so that the curve resembles a spike more than a peak. The DSC of TAGN alone is shown again as a reference for which to compare temperature shifts. This result had previously been observed in another laboratory using a different model of DSC.[11] Since variations in design (different sample containers, variations in purge flow over sample, etc.) can result in differences in pressure and atmosphere composition of the sample surroundings, results on this DSC were obtained for comparison.

The MS results indicate that the initial decomposition at mild temperatures (190-220°C) is due to evolution of $NH_3$ and $N_2$ and not to loss of $HNO_3$. The enrichment of C and $O_2$ in the residue (and corresponding loss of $N_2$ and $H_2$) as well as the increase in nitrate bands in the residue as seen by IR support this. It is interesting to note that the residue is typically a light to medium pink color (the pink color increasing with the degree of decomposition) and that the sublimate was intensely red. If these pink decomposition products are related to the pink color seen in impure TAGN, then these results also do not support the hypotheseis that the impurity in pink TAGN forms by initial cleavage and loss of $HNO_3$. Each of these samples showed an enrichment of nitrate.

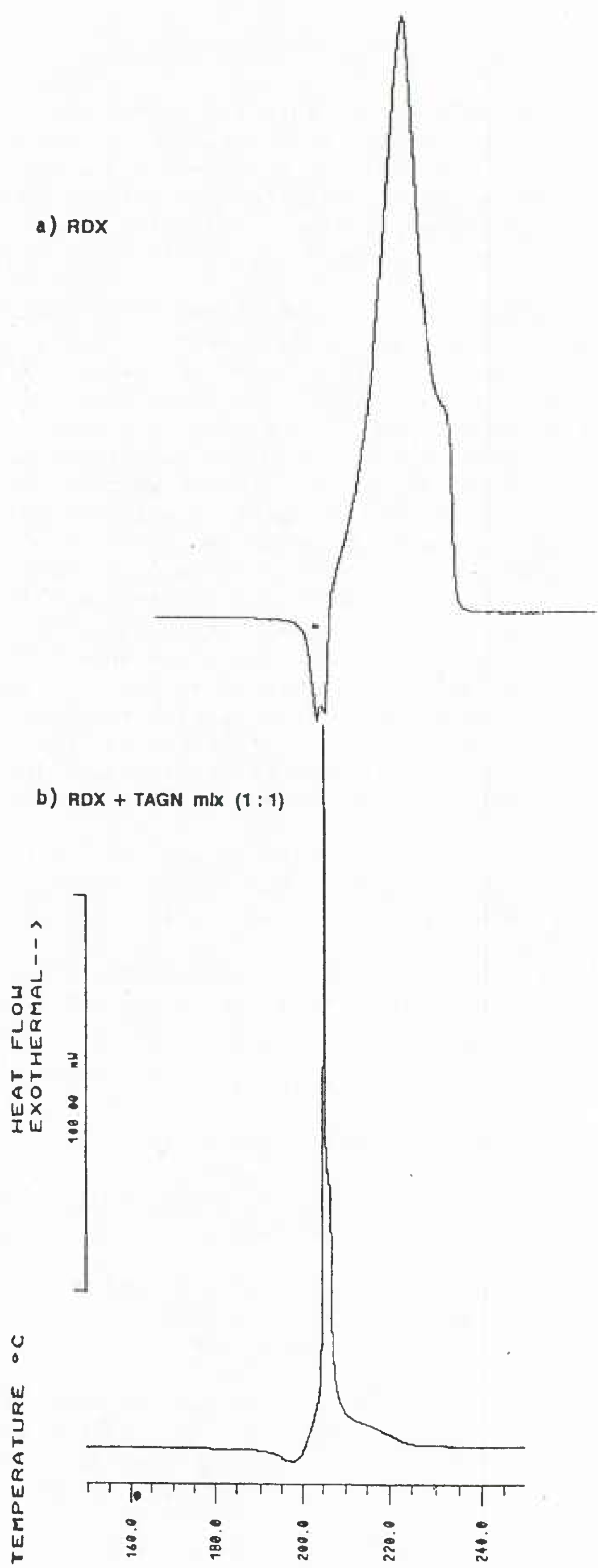


Figure 7. DSC Analysis of (a) RDX and (b) a 1:1 Mixture of RDX and TAGN

## VIII. THIN LAYER CHROMATOGRAPHY

It is desirable to be able to separate the decomposition products of TAGN and a partial separation has recently been achieved. Preliminary attempts to separate the components using normal and both C-18 and amino-substituted reverse phase HPLC failed and thus the faster TLC method was used to select appropriate conditions for the separation. Standards used in the separation are the same as those used as IR references, namely DAGN, AGN, and GN.

A chromatogram showing the separation of two decomposition residues and the standards is given in Figure 8. It is seen that TAGN is the least retained by the plate, followed by DAGN + AGN; GN travels only a small distance with the solvent and is found on the lower portion of the TLC plate. The residue from TAGN thermal decomposition consists of a large amount of DAGN, smaller quantities of AGN and TAGN and no detectable GN. In addition, three components (at least) are present which elute between AGN and DAGN, one of which is quite streaked, possibly due to a significant difference in polarity or functional groups relative to the other samples so that it does not chromatograph as nicely as the others. It is also quite probable that the unknown spots are due to components which are intermediate in size between AGN and DAGN. Considering the structure of TAGN, this may indicate that the bonds between C and N have been broken in these cases, because if only N-N bonds are broken, most probably DAGN, AGN or GN would be formed. Infrared analysis of the spots was attempted. The material from several runs was pooled and extracted and analyzed on a salt plate. Unfortunately, not enough sample was recovered to obtain a spectrum. Reflectance or microsampling attachments were not available but might enable an identification of the sample.

The TLC results support those from the MS and CHN and O analyses, since DAGN and AGN shown here to be present in the residue would be formed from evolution of $NH_3$ and one would expect a relative loss of $N_2$ and enrichment of C. Also, in the infrared spectra of DAGN and AGN the nitrate band is larger relative to the C-N band; their presence or seen in the TLC results supports the finding of a larger nitrate band (relative to the C-N band) in the residue than in the TAGN.

## IX. CONCLUSIONS

The results of the thermal decomposition of TAGN are as follows:

1. Measurement of the evolved gases by MS has indicated that TAGN loses $NH_3$ and $N_2$ as its early decomposition products.

2. The MS results were supported by C, H, N, and O analysis of the residue; it was found to be enriched in carbon and oxygen and showed a loss of nitrogen and hydrogen, compared to unheated TAGN.

3. Infrared analysis indicated that the residue from TAGN heated between 210 and 220°C is probably a mixture consisting of DAGN and AGN, based on band frequencies and relative intensities. A substance which condensed on the walls of the container during heating was identified as DAGN.

4. TLC analysis of the TAGN residue also indicated the presence of DAGN and AGN, as well as at least three other unknown products.

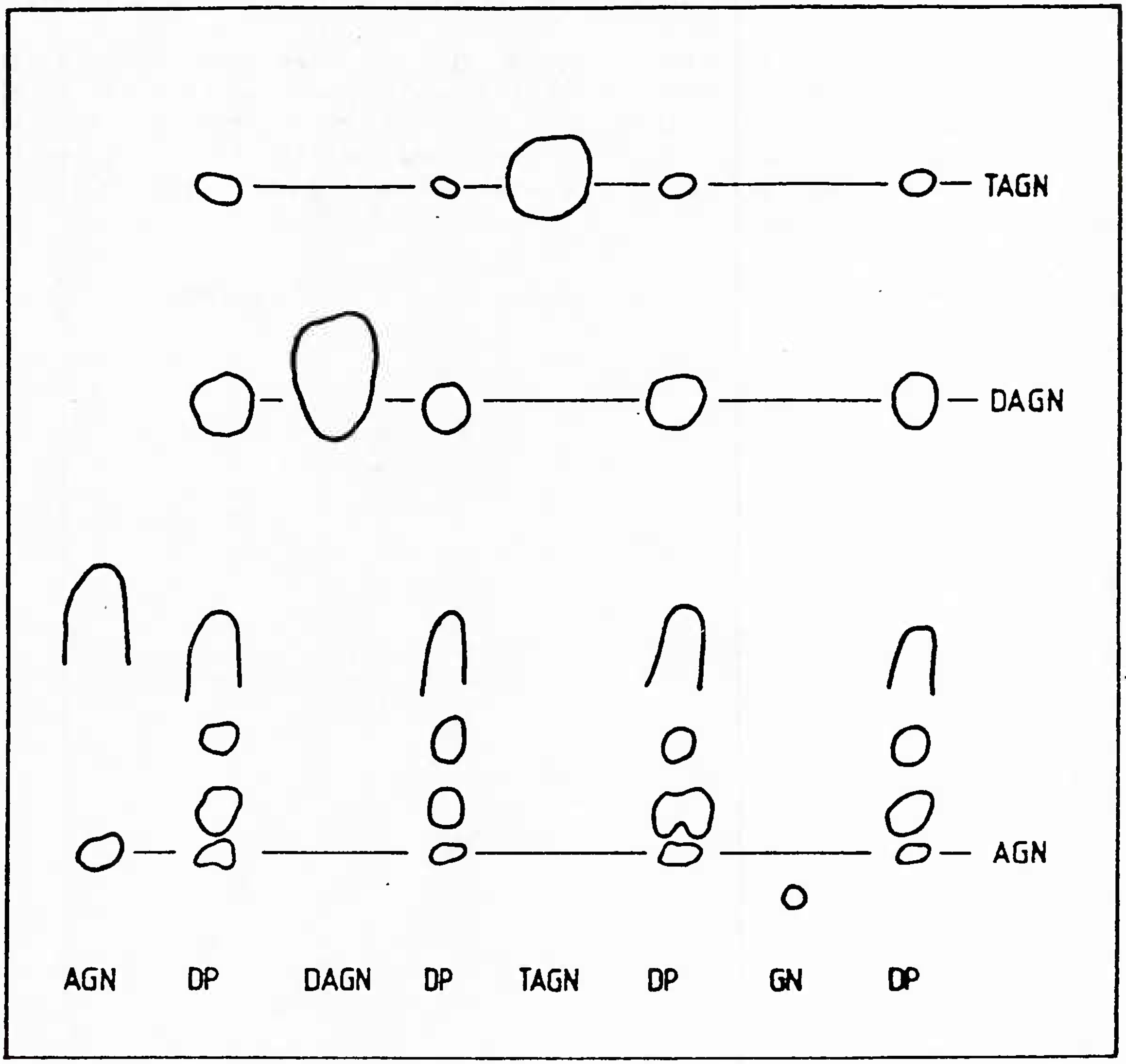


Figure 8. Thin Layer Chromatographic Separation of TAGN Decomposition Products

5. DSC analysis showed that the $NH_3$ and $N_2$ evolution is the first major exothermic reaction in TAGN decomposition.

In addition, a pink TAGN was investigated. The FTIR analysis indicated that the TAG portion of the molecule had not undergone decomposition and the only difference between the pink TAGN and pure TAGN was the greater intensity of the nitrate band in the pink sample. A sample of deep red TAGN crystals was prepared and its spectrum was also identical to TAGN with the exception of an even greater nitrate band intensity (relative) than was seen in the pink species. The source of this strong nitrate band is under investigation.

In conclusion, the results of MS, IR, TLC, and elemental analyses agree and indicate that TAGN decomposes under the conditions used to yield mainly $NH_3$ and $N_2$ and a residue containing DAGN, AGN and other products. No evidence for GN formation was obtained. DSC results show that the $NH_3$ and $N_2$ evolution is the first exothermic step in the decomposition of TAGN. Pink TAGN is possibly related to an enrichment of nitrate.

## REFERENCES


1. H.W. Voight and J.P. Picard, "RDX/TAGN Propellants of Increased Heat Stability and Safe Pressure Exponent," Third International Gun Propellant Symposium, Vol. IV, pp. 248-267, 1984.

2. O.K. Heiney, "Nitramine Gun Propellant for 30MM Gau-8 Application," Third International Gun Propellant Symposium, Vol. IV, pp. 209-225, 1984.

3. N.F. Stanley, "Energetic, Cool-Burning Gun Propellants," Air Force Armament Laboratory, AFATL-TR-78-136, September 1979.

4. J.E. Flanagan, "Advanced Gun Propellant for GAU-8", Proceedings of 11th JANNAF Combustion Meeting, CPIA Publ. 261, Vol. I, pp. 285-291, December 1974.

5. F. Schedlbauer, "Investigation of the Combustion Behavior of LOVA-Propellants," ADPA Proceedings, Long Beach CA, 1986.

6. D. Muller, Fraunhofer Institute (ICT), West Germany, personal communication.

7. Y. Oyumi and T.B. Brill, "Thermal Decomposition of Energetic Materials. 6. Solid-Phase Transitions and the Decomposition of 1,2,3-Triaminoquanidinium Nitrate," J. Phys. Chem., Vol. 89, pp. 4325-4329, 1985.

8. C.W. Fong, "The Formation of Colored Impurities in Triaminoguanidine Nitrate and Related Incompatibility Problems in Gun Propellants," Air Force Armament Laboratory, AFATL-TR-81-101, August 1982.

9. A.J. Bracuti, "The Crystal Structure Determination of Triaminoguanidine Nitrate," ARDC, ARLCD-TR-78050, 1979.

10. F. Volk, Fraunhofer Institute fur Treib und Explosivstoffe, ICT, Karlsruhe, West Germany, private communication.

11. P.J. Kaste and L.J. Decker, BRL Technical Report in progress.

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